



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of:

SATO et al.

U. S. Patent Application No. 09/987,463

Group Art Unit: 1765

Filed: November 14, 2001

Examiner: KUNEMUND. ROBERT M

For: QUARTZ GLASS CRUCIBLE FOR PULLING UP SILICON SINGLE  
CRYSTAL AND PRODUCTION METHOD THEREFORE

**DECLARATION UNDER 37 CFR § 1.132**

Honorable Commissioner of Patents and Trademarks

Washington, D. C. 20231

Sir:

I, Tatsuhiro SATO, a citizen of Japan hereby declare that:

1. I am one of the inventors of the above-identified application and am fully familiar with the subject matter thereof, as well as the references relied upon by the Examiner.

2. I understand that the Examiner in position that since the combined prior art uses process similar to that claimed the crucible would inherently have the same properties after pulling.

3. I supervised the following additional comparative examples of the different results using a different gas content, an electric power and a diameter from the claimed thereof.

(Comparative Example 2) A quartz glass crucible 22 inches in diameter was produced in accordance with the below described procedure using the apparatus shown in FIG. 1. Natural quartz glass powder in the range of 50 to 500  $\mu\text{m}$  in particle diameter was fed into a mold (of a rotary type) 560 mm in inner diameter, rotating at 100 rpm, and piled up on the inner surface to a thickness of 30 mm in a uniform manner and then not only was the piled up quartz powder molten by heating from the interior of the mold by means of arc discharge but at the same time, synthetic quartz powder, which had a particle diameter in the range of 50 to 300  $\mu\text{m}$ ; an OH group concentration of 100 ppm; a gas content of 60  $\mu\text{l/g}$  including  $\text{CO}$ ,  $\text{CO}_2$  and  $\text{CH}_4$  of a total 25 % by volume, which had been quantified by means of a temperature programmed gas analysis from 0 up to 1000 degrees, was continuously fed from above at a feed rate of 100 g/min at a position spaced from the center of arc by a horizontal distance of 100 mm, under other conditions that the arc was held at a height of 500 mm above the inner surface of the bottom which was formed by melting; the arc was moved sideways in a horizontal plane from the central position of the mold; and an amount of arc heat was provided through the process at an electric power of 500 kw under conditions of a DC current of 1500 A and a voltage of 200 V, with the result that a transparent glass layer was formed across the entire inner surface to a thickness of 1 to 3 mm.

After completion of the quartz glass crucible, a gas content in the transparent layer 1 mm in depth from the inner surface was measured by means of the temperature programmed gas analysis in the range of 0 to 1000 degrees and as a result, the gas content was measured to be equal to or less than 3 $\mu\text{l/g}$ . Further, the maximum bubble diameter of a inner surface layer 1 mm in depth of the quartz glass crucible was equal to or less than 0.3 mm, a total bubble sectional area was measured to be equal to or less than 20 % and  $\text{CO}$ ,  $\text{CO}_2$  and  $\text{CH}_4$  of a total 25 % by volume was included in bubbles. A microphotograph of a sectional structure of a

piece 1 mm in thickness sampled from a round portion of the quartz glass crucible produced in the example is shown in FIG. 4.

Another quartz glass crucible produced in the example was used to melt polysilicon charged therein and a silicon single crystal was pulled up in the passage of 100 hr under a degree of vacuum of 20 mmb and the silicon single crystal went into a disorder in the course of pulling up. The transparent layer of the inner surface of the quartz glass crucible after use was observed and many bubbles of 0.8 mm were found in a layer 0.5 mm in depth from the inner surface with some bubbles having burst after reaching the inner surface from inside during the pulling up and a total bubble sectional area was 60 %. A piece 1 mm in thickness was sampled from a round portion of the quartz glass crucible after use and a microphotograph of a sectional structure was taken, which is shown in FIG. 5.

(Comparative Example 3) A quartz glass crucible 30 inches in diameter was produced in accordance with the below described procedure using the apparatus shown in FIG. 1. Natural quartz glass powder in the range of 50 to 500  $\mu\text{m}$  in particle diameter was fed into a mold (of a rotary type) 560 mm in inner diameter, rotating at 100 rpm, and piled up on the inner surface to a thickness of 30 mm in a uniform manner and then not only was the piled up quartz powder molten by heating from the interior of the mold by means of arc discharge but at the same time, synthetic quartz powder, which had a particle diameter in the range of 50 to 300  $\mu\text{m}$ ; an OH group concentration of 100 ppm; a gas content of 60  $\mu\text{l/g}$  including  $\text{CO}$ ,  $\text{CO}_2$  and  $\text{CH}_4$  of a total 25 % by volume, which had been quantified by means of a temperature programmed gas analysis from 0 up to 1000 degrees, was continuously fed from above at a feed rate of 100 g/min at a position spaced from the center of arc by a horizontal distance of 100 mm, under other conditions that the arc was held at a height of 500 mm above the inner surface of the bottom which was formed by melting; the arc was moved sideways in a horizontal plane from the central position

of the mold; and an amount of arc heat was provided through the process at an electric power of 750 kw under conditions of a DC current of 1500 A and a voltage of 200 V, with the result that a transparent glass layer was formed across the entire inner surface to a thickness of 1 to 3 mm.

After completion of the quartz glass crucible, a gas content in the transparent layer 1 mm in depth from the inner surface was measured by means of the temperature programmed gas analysis in the range of 0 to 1000 degrees and as a result, the gas content was measured to be equal to or less than  $3\mu\text{l/g}$ . Further, the maximum bubble diameter of a inner surface layer 1 mm in depth of the quartz glass crucible was equal to or less than 0.3 mm, a total bubble sectional area was measured to be equal to or less than 20 % and CO, CO<sub>2</sub> and CH<sub>4</sub> of a total 25 % by volume was included in bubbles. A microphotograph of a sectional structure of a piece 1 mm in thickness sampled from a round portion of the quartz glass crucible produced in the example is shown in FIG. 4.

Another quartz glass crucible produced in the example was used to melt polysilicon charged therein and a silicon single crystal was pulled up in the passage of 100 hr under a degree of vacuum of 20 mmb and the silicon single crystal went into a disorder in the course of pulling up. The transparent layer of the inner surface of the quartz glass crucible after use was observed and many bubbles of 0.8 mm were found in a layer 0.5 mm in depth from the inner surface with some bubbles having burst after reaching the inner surface from inside during the pulling up and a total bubble sectional area was 60 %. A piece 1 mm in thickness was sampled from a round portion of the quartz glass crucible after use and a microphotograph of a sectional structure was taken, which is shown in FIG. 5.

4. In considering the results of these experiments and based on my knowledge of the state of the prior art, it is my opinion that the invention defined by the claims of the above identified patent application represents an unobvious

departure from the prior art because the advantages achieved in the present invention would not occur if the variables are changed from those recited in the present claims.

5. I, Tatsuhiro SATO, the undersigned declarant declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001, of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 18th day of February, 2004.

Tatsuhiro Sato

Tatsuhiro SATO